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1 Sediment fingerprinting as a tool to identify temporal and spatial variability of sediment sources and
2 transport pathways in agricultural catchments

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Abstract

Management strategies to reduce soil loss and sediment delivery from agricultural land requires an empirical understanding of sediment sources. Sediment fingerprinting is a technique to apportion sources to a downstream sediment sample which, when applied at high spatial and temporal resolutions, can offer insights into catchment sediment dynamics. However, developing an overarching tool can be hindered due to indeterminate interactions such as, for example, landuse, soil and geological conditions and multiple sediment source pressures. To address this, a multi-proxy sediment fingerprinting approach was used in three catchment observatories in Ireland, characterised and referred to by their predominant soil drainage and land use characteristics: poorly-drained grassland, well-drained arable and moderately-drained arable. Potential sediment source groups: channels, field topsoils, and roads, were sampled. Target sediment samples were collected from six sites within each catchment over approximately two-years from May 2012 to May 2014. Geochemical, mineral magnetic and radionuclide tracers were measured in source and target sediment samples and, following justified tracer selection, source proportions were estimated using an uncertainty inclusive un-mixing model. Overall, the poorly-, well- and moderately-drained catchments exported 828, 421 and 619 tonnes, respectively ($36, 19$ and $33 \text{ t km}^{-2} \text{ yr}^{-1}$). Estimated source contributions from channel, field topsoil and road groups were overall, 67%, 27% and 4% in the poorly-drained grassland, 53%, 24% and 24% in the well-drained arable and 9%, 82% and 8% in the moderately-drained arable catchment outlets. Sub-catchment source estimates were generally consistent with the outlet over space and time. Short-term activation of previously unidentified transport pathways were detected, for example, field sources transported by the road network in the well-drained catchment. In catchments with high hydrological surface connectivity (moderate and poor soil drainage), exposed soils were most sensitive to soil erosion and sediment delivery. Where groundcover is maintained on these soils, sediment connectivity was diminished and flow energy is transferred to the stream network where channel bank erosion increased. In the well-drained catchment, sub-surface flow pathways dominated and consequently channel sources, broadly representative of subsoil characteristics, were the largest sediment source. Sediment connectivity contrasted in the studied agricultural catchments according to

41 source availability, and erosion, transport and delivery processes. Effective sediment management
42 strategies in intensive and intensifying agricultural catchments must consider sediment loss risk
43 resulting from catchment specific sediment connectivity and emphasise mitigation strategies
44 accordingly.

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46 Keywords:, soil erosion, water quality, agriculture, catchment management, connectivity

1 Introduction

Intensive agricultural systems, resulting in enhanced soil erosion and sediment delivery can pose risks to aquatic ecosystems such as rivers and lakes (Collins and Zhang, 2016; Borelli et al., 2017; Tiecher et al., 2017; Vanwalleggem et al., 2017). In agricultural catchments, fluctuations in groundcover due to arable cultivation or livestock poaching (soil structural damage due to animal trampling) exposes the soil surface to erosional processes, thereby increasing their sensitivity to soil erosion and subsequent sediment loss (Haygarth et al., 2006). Land management, such as installation of artificial drainage, promotes aeration and alleviates excess soil moisture, thereby increasing the productivity of soils (Ibrahim et al., 2013). This also increases the efficiency of hydrological transfers from hillslopes to channels and runoff ratios (Shore et al., 2013). Moreover, landscape modifications interact with local heterogeneous catchment attributes (landscape position, slope, soil drainage, antecedent conditions) and rainfall to alter the distribution of soil erosion and sediment delivery, i.e., sediment connectivity (Sherriff et al., 2016).

In waterbodies, augmented supply of sediments to the channel bed can cause degradation of aquatic habitats resulting in reduced species diversity, as specifically noted in Ireland (Davis et al., 2018) and France (Descloux et al., 2013), and extensively reviewed throughout the world by Kjelland et al. (2015). High suspended sediment concentrations in aquatic ecosystems, for example following rainfall events, also reduce habitat quality for example resulting in increased drifting of invertebrates, commonly used as bioindicators (Kjelland et al., 2015; Béjar et al., 2017). Overall, reduction of ecological diversity challenges the achievement of ecologically “good” status as required under the EU Water Framework Directive (WFD; 2000/60/EC, Official Journal of the European Communities, 2000). Catchment management strategies require identification of sediment sources and an

understanding of the spatial and temporal dynamics of physical processes to cost-effectively target and reduce on-farm soil loss and off-farm downstream sediment supply (Walling et al., 2008).

There are difficulties in fully defining catchment sediment risks and monitoring mitigations. Firstly, auditing individual soil erosion and sediment storage components into a catchment sediment budget demands considerable investigation time and resources (Walling and Collins, 2008). Secondly, establishing an evidence-base, relating specific agricultural practices to different sediment sources and delivery pathway fluctuations over multiple seasons, requires a representatively long study period with observations at an appropriate resolution (Sherriff et al., 2015a). Alternative catchment-scale techniques such as sediment fingerprinting have, therefore, emerged as an effective management tool in river catchments (Gruszowski et al., 2003, Rowan et al., 2012; Thompson et al., 2013; Lamba et al., 2015).

The sediment fingerprinting approach assumes that physico-chemical properties of minerogenic sediment, the inorganic component, can be conserved along a transport pathway, providing the numerical basis to ‘unmix’ the the composite-signatures of suspended sediments samples during flood events or from sediment stores such as channel beds, floodplains and lakes (Pulley et al., 2015) and to apportion the relative contribution to their respective upstream sources (Haddadchi et al., 2013). The upstream catchment is subdivided into potential sources (or source group types) that can be distinguished by their properties, for example, according to land use (Gruszowski et al., 2003; Blake et al., 2012), lithology (Collins et al., 1998), or erosional processes (Fox and Papandicolou, 2008).

Sediment tracers typically employed include geochemistry, mineral magnetism and environmental radionuclides and are potentially numerous considering the availability of modern analytical equipment (Pulley and Rowntree, 2016). However, selected tracers must be conservative (resistant to chemical transformation) and their environmental significance justified in terms of the ability to discriminate between environmentally relevant sources (Koiter et al., 2013). Furthermore, it is assumed the impact of physical processes (erosion, transport, deposition, and re-entrainment) on tracer concentrations due to particle size selectivity and organic matter variation, can be numerically corrected. Simple correction factors are commonly used (Collins et al., 2001), but the appropriateness of these is now disputed (Smith and Blake, 2014) and more refined approaches involving particle size fractionation are an alternative (Motha et al., 2004; Small et al., 2004). Sediment contributions from each source are determined using statistically-based un-mixing algorithms, frequently accompanied by uncertainty estimates (Franks and Rowan, 2000; Sherriff et al., 2015b).

Sediment fingerprinting studies have been applied across a range of scales designed to explore the variability of sediment sources to a single area of impact, e.g., lake, degraded gravel habitat, or streams (Pulley et al., 2015). Particular advances have included assessing high-resolution temporal changes in sediment sources and investigations across hydrological regimes (Cooper and Krueger, 2017; Rose et al., 2018; Tiecher et al., 2018). However, the negative impacts of excessive sediment transport and/or deposition may extend far upstream of a catchment outlet (Fryirs et al., 2007). As such, there is a need to define sub-catchment variability of sediment sources to overcome the indeterminate potential of interacting land use, soil/geology and source variability issues that exist with sediment dynamics in catchments. This can facilitate interrogation of catchment hydrological and sediment

connectivity processes inferred at the catchment scale, and how they relate to the spatially heterogeneous pattern of land use (e.g., crop type, animal grazing) and land management (e.g., riparian vegetation, arrangement of farm tracks – Sherriff et al., 2016).

Correct identification of sediment sources and disentangling the processes controlling soil erosion, sediment entrainment, transfer and deposition will provide an evidence base for application of targeted on- and off-farm sediment management strategies (Rowan et al., 2012). This is particularly important in catchments with contrasting physical and agricultural land management characteristics where targeted strategies may be different (Sherriff et al., 2016). Appropriate source-based mitigation measures are necessary to prevent off-farm nutrient and sediment supply downstream (Evrard et al., 2007; Deasy et al., 2010). Successful application of suitable mitigation measures are essential to reduce on-farm nutrients and soil losses through the preservation of chemical, physical and biological soil quality (Cerdà et al., 2017). These are important considerations to reduce the environmental impact of intensive agriculture and also to offset the likely changes occurring as land becomes more intensively managed. Increased or changing land use (crop types, animal numbers), soil drainage and increased machine trafficking are all likely to occur under scenarios of agricultural intensification in Ireland, Europe and worldwide (Ewert et al., 2005; Coyle et al., 2016; Teshager et al., 2016).

The overall aim of this study was to use sediment fingerprinting to define the spatial and temporal variability of sediment sources of instream sediments in intensive or intensifying agricultural catchments. The sediment fingerprinting methodology used a multi-proxy suite of environmental radionuclides, geochemistry and mineral magnetism within a statistically based un-mixing framework. This method was applied in three catchment observatories in

146 order to fulfil two objectives. Firstly, to assess relative magnitudes of sediment sources
147 between catchments with contrasting land use and dominant soil drainage characteristics.
148 Secondly, to assess the spatial and temporal variability of sediment sources within each
149 catchment. This analysis was used to recommend catchment and source specific measures to
150 reduce the soil and sediment loss from land.

2 Methods

2.1 Catchment observatories

Sediment fingerprinting studies were focussed on three lowland intensive agricultural catchments in Ireland. Consistent with Sherriff et al. (2016), these catchments are named according to their dominant soil drainage and predominant land use types; poorly-drained grassland, well-drained arable and moderately-drained arable (Fig. 1).

The poorly-drained grassland catchment (11.0 km²) with median slopes of 3°, is located in south-east Ireland. It is geologically permeable owing to Ordovician Volcanics and metasediments of the Campile formation (Tietzsch-Tyler et al., 1994). However, overlying Groundwater Gley soils in the lowlands (Luvic Stagnosol, sandy loam - World Reference Base classification for soils, Creamer, 2014; 2016), influenced by the Irish Sea till subsoil, dominate the catchment area and consequently impede drainage once the well-drained upper horizons are saturated. Well-drained Brown Earths (Haplic Cambisol, loam) are found in limited areas of the uplands but overall surface flow hydrological pathways dominate in this catchment (Mellander et al., 2012). Consequently, artificial surface (open drainage ditches) and sub-surface (piped) drainage networks are widespread across the landscape. Grassland agriculture for sheep, beef and dairy grazing (77%) is the predominant land use, with arable crops (12%) primarily spring cereals contained to upland well-drained soils.

The well-drained arable catchment (11.2 km²) with median slopes of 4° is located in south-east Ireland. The catchment is geologically composed of slate and siltstones of the Oaklands Formation (Tietzsch-Tyler et al., 1994). This provides poor primary permeability but secondary productivity creates opportunities for water transfer through fissure flow. Overlying soils are predominantly well-drained Brown Earths (Haplic Cambisol, loam) with

limited areas of poorly-drained Groundwater Gleys (Haplic Gleysol, clay loam) in the eastern stream corridor leading to a dominance of sub-surface hydrological pathways in this catchment. Artificial drainage networks are limited to the poorly-drained catchment areas which are primarily utilised for permanent grassland (39% of catchment) for beef cattle and sheep grazing. Arable crops (54% of catchment), mostly spring barley, are supported by well-drained soils which undergo limited rotation between years.

The moderately-drained arable catchment (9.5 km²) is located in north-east Ireland and has median slopes of 3°. Geologically, it is characterised by calcareous greywacke sandstones and banded mudstones (McConnell et al., 2001) which results in a poorly productive aquifer (Mellander et al., 2012). The overlying soils are categorised as moderate and poorly-drained dominated by A-horizon loams and B-horizon clay loams underlain by fine till containing siliceous stones and fluvioglacial sediments in the channel corridor (Haplic Cambisols, loam and Gleyic Fluvisol, silt). Surface hydrological pathways dominate in the catchment but a greater sub-surface hydrological influence has been detected during winter months (Melland et al., 2012; Mellander et al., 2012). Artificial drainage networks are dominant in poorly-drained areas but generally widespread across the catchment. A large proportion of mixed arable crops (42%) including winter-sown cereals, maize, potatoes among others are supported in the catchment and the other land use is permanent grassland (48%) to support grazing of sheep and cattle for dairy and beef.

2.2 Fingerprinting sample collection

Reconnaissance surveys identified six primary potential sediment sources in the study catchments: grassland topsoils, arable topsoils, damaged road verges, farm tracks, eroding channel banks and eroding ditch banks, which were sampled for analysis. Field sample

locations were randomly selected using a spatial dataset of fields stratified by their general land use, grassland or arable. The proportion of fields sampled from each land use approximately reflected the overall proportion of that land use contained within each catchment. Each field topsoil sample (0-5 cm) was collected with an auger and comprised a composite of multiple sub-samples within each field. In the poorly-drained grass, well-drained arable and moderately-drained arable catchments, 22 fields (grassland n=16; arable n=6), 24 fields (grassland n=9, arable n=16) and 30 fields (grassland n=19, arable n=11) were sampled, respectively.

Channel and drainage ditches were sampled during winter 2013 and 2014 when vegetation cover was low. Samples targeted actively eroding areas from which a composite sample of vertical bank section was collected with a trowel (from a stream reach no greater than the adjacent field width), with opposite river banks collected separately. In total, 62 channel samples were collected from the poorly-drained grass catchment, 15 from the well-drained arable catchment and 14 from the moderately-drained arable catchment. Time constraints associated with radionuclide isotope analysis resulted in analysis of a randomly selected subset of 30 samples (from the 62) which were deemed appropriate, based on the number of samples used to represent other potential sources, to characterise the channel sediment source in the poorly-drained grass catchment (Fig. 1). Active erosion of ditch (open field drain) channels was observed only in the poorly-drained grassland catchment (n=4) and was sampled consistently with channel banks.

Surface scrapings of damaged road verges and farm tracks were collected with a trowel along a maximum road or track length of approximately 200 m and compositing, where relevant, both sides of the road into one sample (Fig. 1). Twenty-three, eight and 12 samples were

collected in the poorly-drained grass, well-drained arable and moderately-drained arable catchments, respectively.

River sediment samples were collected using time integrated suspended sediment (TISS) samplers (Phillips et al., 2000) at multiple locations within each study catchment from May 2012 to May 2014 (Fig. 1). The samplers are constructed of a main body (98 mm internal diameter PVC pipe, 1 m length) capped with an upstream facing funnel and a closing downstream cap. Small diameter inlet/outlet tubes (4 mm diameter, approximately 20 cm length) facilitate the flow of water through the sampler. Suspended sediment particles are deposited inside the main body when leaving the inlet tube due to the reduction in velocity coinciding with the increase in diameter. Sediment samples were collected at 6-12 week intervals to assess seasonal changes in sediment sources (Table 1). Missing data were attributed to equipment malfunctions or insufficient sample quantity for analysis (using label locations and sample times in Fig. 1 and Table 1: MD3 – T14, MD1 and MD5 – T10, PD4 – T11, WD4 – T3, WD6 – T7, T8 and T11). Site PD3 was removed following vandalism and a replacement site, PD1, established upstream to coincide with the deployment of samplers on 13/05/2013. At WD5, channel reconfiguration (following T10) resulted in cessation of sample collection at this location. Short term site inaccessibility (due to inundated channels) prevented the collection of the MD3 and WD1-WD5 at the end of T9. These samples were retrieved at the next collection occasion and were therefore representative of the two periods T9 and T10, combined.

2.3 Suspended sediment load estimation

Catchment outlet suspended sediment loads were estimated using turbidity (Solitax, Hach-Lange, Germany) and water level (m) data collected from a vented pressure-transducer (OTT

Orpheus-mini, OTT, Germany) located inside a stilling well at 10-min resolution from May 2012 to May 2014. Turbidity data were converted to suspended sediment concentrations (SSC) using detailed cross-sectional turbidity-SSC calibrations (Sherriff et al., 2015a). The velocity-area rating method for gauging instantaneous discharge was used to calculate discharge over a non-standard Corbett flat-v weir (Corbett Concrete, Cahir, Ireland) using WISKI-SKED software. Meteorological data; 10-min rainfall, air temperature, relative humidity, radiation and wind-speed were collected from lowland weather stations in each catchment (BWS200, Campbell Scientific).

2.4 Laboratory analysis

Geochemical, radionuclide and mineral magnetic analysis was conducted on soil and sediment samples. Geochemical elements (Cd, Co, Cr, Cu, Mn, Ni, Pb and Zn) were analysed using an Agilent ICP-OES (Santa Clara, US) following microwave assisted acid digestion (USEPA, 1996) to obtain total concentrations (mg kg^{-1}). Radionuclide activity mass concentrations (Bq kg^{-1}) of ^{210}Pb , ^{234}Th , ^{235}U , ^{214}Pb , ^{137}Cs , ^{228}Ac , ^{40}K were measured using a low background Ortec HPGe gamma spectrometer detector (Model no. GEM-FX7025-S) after samples were radon-sealed inside 55 mm petri dishes for a minimum of 30 days to determine the unsupported fraction ($^{210}\text{Pb}_{\text{unSUPP}}$), of ^{210}Pb activity (Foster et al., 2007; Rowan et al., 2012). Detector calibration was achieved using a National Physics Laboratory mixed-gamma standard (R08-03) within standardised mass/geometries (1, 5, 10 g). Mineral magnetic measurements, the mass-specific low field susceptibility ($\chi_{\text{LF}} - 10^{-6} \text{ m}^3 \text{ kg}^{-1}$), high field susceptibility ($\chi_{\text{HF}} - 10^{-6} \text{ m}^3 \text{ kg}^{-1}$), frequency-dependent susceptibility ($\%\chi_{\text{FD}}$), anhysteretic remanence magnetisation ($\chi_{\text{ARM}} - 10^{-7} \text{ Am}^2 \text{ kg}^{-1}$), saturation isothermal remanent magnetisation (SIRM at 1 T $- 10^{-5} \text{ Am}^2 \text{ kg}^{-1}$), backfield IRM measurements IRM_{soft} and

bIRM_{hard} were completed, and ratios SIRM/ χ_{LF} , SIRM/ χ_{ARM} , χ_{ARM}/χ_{LF} and the H-ratio ($0.5 * (SIRM - bIRM_{40})$) calculated.

The specific surface area (SSA – $m^2 kg^{-1}$) of soil and sediment samples were measured using a Malvern Mastersizer Hydro 2000G (range 0.02 to 2000 μm) following organic matter removal and chemical/physical dispersion (Fenton et al., 2015). Total carbon (TC) and total organic carbon (TOC – following acid treatment of the inorganic fraction with hydrochloric acid (Massey et al., 2013)) were analysed on a LECO Truspec CN analyser (LECO Corporation, Michigan, USA) as a proxy for organic matter content. Samples were individually corrected for particle size (corrected tracer concentration = measured tracer concentration/SSA – Gruszowski et al., 2003) and organic matter content (corrected tracer concentration = particle size corrected concentration/ % organic carbon).

2.4 Statistical analysis

The capability of an individual tracer to distinguish between sources was assessed using the Kruskal-Wallis test (SPSS v. 22.0; IBM, USA) – $p < 0.05$), with tracers that differed significantly between sources being retained for further analysis. This was followed by pairwise analysis using the Dunn-Bonferroni test (adjusted $p < 0.05$) to determine significant differences between groups. Under spatial interrogation, source sample tracer characteristics did not display sub-catchment trends within each study catchment; therefore, all source samples, regardless of their catchment location, were utilised for un-mixing suspended sediment samples. Non-conservative behaviour of tracers in suspended sediment samples was identified by values exceeding the range defined by source samples were removed from further analysis (Mukundan et al., 2010; Smith and Blake, 2014). Removal of $^{210}Pb_{unsp.}$ and ^{137}Cs where concentrations were 0 $Bq kg^{-1}$ in suspended sediment samples occurred as,

300 although tracers were not non-conservative, their impact on result predictions and uncertainty
301 were considerable, consistent with Sherriff et al. (2015b). All remaining tracers were
302 subsequently interrogated to justify their environmental significance (Koiter et al., 2013).
303 Tracer values were entered into Multiple Discriminant Analysis (MDA) to determine the
304 discrimination capability of the resulting tracer set for each outlet and sub-catchment
305 suspended sediment sample (Table 5). Subsequently, source contributions were un-mixed
306 using the uncertainty inclusive FR2000 model (Franks and Rowan, 2000), with uncertainties
307 determined on probability distributions from the input dataset of tracer values and target
308 sediments. For simplification, the distance between the upper and lower uncertainty values,
309 representing the 95th percentile for each source, were combined for each target sediment
310 sample.

3 Results and discussion

3.1 Tracer selection

The six sampled source groups could not be discriminated using MDA when deploying the full tracer arrays in all three study catchments (Yu and Rhoads, 2018). However, three composite ‘parent’ groups were identified and attributed to similar soil loss processes:

- Channels: comprising channel banks and ditches
- Field topsoils: comprising arable and grassland topsoils
- Roads: comprising road verges and tracks.

Source group tracer summaries are shown in Table 2 – poorly-drained grassland, Table 3 – well-drained arable and Table 4 – moderately-drained arable and hereafter the use of ‘channel’, ‘field’ and ‘road’ refers to parent groups unless specified. Tracers that were statistically non-significant between at least two sources (Kruskal Wallis $p > 0.05$) were removed from analysis (Tables 2-4).

In all catchments, road samples were generally elevated compared to other sources for mineral magnetic tracers (χ_{LF} , χ_{HF} , χ_{ARM} , SIRM) and/or metallic elements Cu, Pb and Zn, reflecting possible inputs from vehicle exhausts (Rose et al., 2018). This trend was not reflected in the well-drained arable catchment likely due to greater ferrimagnetic minerals (high IRM_{soft}) due to iron-rich geology. Surficial sources were well defined by ^{137}Cs in the poorly-drained grassland (Table 2) and well-drained arable catchments (Table 3). In the moderately-drained arable catchment (Table 4), however, this tracer failed to distinguish between any sources.

All catchments showed higher $\% \chi_{FD}$ in field topsoils soils than channel sources. Maher and Taylor (1988) attributed elevated $\% \chi_{FD}$ in topsoils to the production of magnetite grain

coatings in the surface of poorly-drained soils. Similarly elevated values but less group variability in the well-drained catchment was attributed to higher background ferromagnetic material rather than that produced in-situ. Higher concentrations of Cu and Ni in channel sources compared to topsoils were explained in the poorly-drained grassland and well-drained catchments by reduced weathering (and therefore depletion of element concentration) relative to topsoils (Smith and Blake, 2014). Soil heterogeneity prevented this trend in the moderately-drained arable catchment. Channel samples could be considered a good representation of other sub-surface sources such as drains, gullies and tracks which lose subsoil material (Collins et al., 2010; Cooper and Krueger, 2017). In the poorly-drained grassland catchment, a thick and low-permeability marine clay subsoil present at 1.5 – 2 m (below surface) was successfully characterised in this source group due to occasional exposure at the base of channel bank sections (Mellander et al., 2015).

The suspended sediment samples showed consistent tracer non-conservativeness (more than 36% of samples within a catchment) in two tracers, Cd and Mn, from the poorly-drained grassland catchment; and four tracers, SIRM/ χ_{ARM} , Cd, Cr, and Mn from the moderately-drained arable catchment which were consequently removed from analysis. Removal of non-conservative tracers in individual suspended sediment samples were similarly removed from their associated source dataset. This did not impact significantly on the cumulative discriminatory power as determined by the MDA. The source groupings of the maximal tracer set (Fig. 2) and the distribution of discriminatory power for all suspended sediment samplers (Table 5) qualified against other acceptable values reported elsewhere (Lamba et al., 2015; Theuring et al., 2015).

3.2 Catchment outlet export and sediment source predictions over time

Total suspended sediment export over the study period was 828, 619 and 421 tonnes in the poorly-drained grassland, well-drained arable and moderately-drained arable catchments, respectively (equivalent to 36, 19 and 33 t km⁻² yr⁻¹, respectively). These were higher than the longer-term average yields (hydrological years 2009-2012) previously measured, 25, 12 and 24 t km⁻² yr⁻¹ in the poorly-drained grassland, well-drained arable and moderately-drained arable catchments, respectively, but did not exceed the maximum annual sediment yields (Sherriff et al., 2015a).

Load specific un-mixing using median predictions indicated channels were the dominant sediment sources in the poorly-drained grassland catchment outlet samples (PD6 – Fig. 3a) which overall accounted for 67% (range 42-77%), or 554 t, of the suspended sediment load (SSL). This confirms previous studies suggesting that proximal sediment sources, likely channel banks, were primary contributors to the SSL here (Sherriff et al., 2016). Field topsoils overall contributed 27% (range 19-49%) to the SSL and did not increase with greater sediment export (Sep-12 to Apr-13 and Dec-13 to Mar-14) confirming that, despite good connectivity, hillslope sediment loss risk was largely reduced by permanent pasture groundcover. Roads were a negligible sediment source in this catchment (total 4%, range 1-17%) related to their distal location relative to the stream network. Additionally, roads were frequently bordered by road-side ditches designed to divert and store surface water. Previous analysis in these catchments has indicated that disconnecting ditches from the stream network provides a sediment sink to prevent subsequent transport to watercourses (Shore et al., 2013).

Sediment sources were predominantly channel derived at the well-drained arable catchment outlet – WD6 (53%, range 0-69%), with smaller proportions attributed to field topsoils (24%, range 9-40%) and roads (24%, range 18-47%) over the study period (Fig. 3b). Higher average

sediment exports (greater than $\sim 2 \text{ t day}^{-1}$) were less frequent compared to the other catchments due to the less-flashy hydrological response and dependence of event-scale rainfall characteristics on sediment connectivity (Sherriff et al., 2016). The stream network is largely (approximately 66%) contained by a woodland riparian corridor thereby stabilising channel bank soils through root networks and reducing the potential for bank erosion (Polvi et al., 2014). Despite this, observed and reported representative channel composite sources are from localised drainage programmes (two drainage projects occurred during January 2014 on the north-south tributary with banks without woodland), subsurface sediment sources to tile drain flow (e.g. Deasy et al. 2009), and other efficient subsurface hydrological pathways supported by fractured bedrock (Warsta et al., 2013; Sherriff et al., 2016).

Field topsoils dominated sediment sources in the moderately-drained arable catchment, accounting for 82% of the total yield (range 59-93%) with 9% attributed to channels (range 0-17%) and 8% to roads (range 0-23%) over the study period (Fig. 3c). Foucher et al. (2014) similarly reported the dominance of surface sources in a row crop arable catchment in France with widespread sub-surface drainage. Previous inferences using hysteresis analysis (Sherriff et al., 2016) assigned sediment export risk to bare arable fields with good hillslope connectivity (resulting from predominately moderately- and poorly-drained soils), which these data confirm. Similarly to the well-drained catchment, increased bank stabilisation from the extensive root networks of riparian woodland likely reduced the risk of bank erosion for much of the stream network.

3.3 Sub-catchment sediment sources and uncertainty over space and time

In all study catchments, sub-catchment sediment sources were generally consistent with outlet predictions (Fig. 4). This was most evident in the poorly-drained grassland catchment

whereby channels were consistently the dominant sources throughout the catchment. This indicates the processes of soil erosion and sediment transfer were consistent. Higher field topsoil contributions occurred on average in the lower poorly-drained grassland catchment sites (PD4, PD5 and PD6) indicating that arable topsoils primarily located on well-drained soils in the upper catchment were not contributing significantly. There were some elevated field contributions in samples T9 and T10 but low catchment outlet sediment export here suggests minor impacts on the load particularly in T9 where result predictions were associated with greater uncertainty (Fig. 5).

The well-drained arable sub-catchment samples showed, similarly to the outlet, channel sources dominated. However, greater field topsoil contributions occurred during periods of increased SSL (T4, T5 and T12) reflecting greater sediment connectivity during wetter periods. Sediment source estimations at sites WD1 and WD4, which drain the north-south tributary, were consistently elevated for field topsoils relative to other sites. This suggests sediment connectivity was greater here, where hillslopes were steeper with a narrow stream corridor and a reduced riparian woodland cover (Wischmeier and Smith, 1978; Gurnell, 2014). Increased field topsoils here coincided with greater road contributions suggesting runoff over impermeable surfaces is an important delivery mechanism for field eroded soils. Road verges, in addition would contribute as a separate sediment source (Gruszowski et al., 2003). At WD6 during T13, channel sources were negligible and showed inconsistencies over time (WD6) and space (T13 samples) – but with high uncertainty in the predictions (Fig. 5).

In the moderately-drained arable catchment, field topsoils contributed on average greater than 80% across all sub-catchment sites over time and showed less variability relative to other study catchments. The availability of sediment sources and surficial mechanisms of soil

erosion, sediment transport and delivery were, therefore, consistent across the catchment and over time (Sherriff et al., 2016). Similarly to the downstream sites in the well-drained arable catchment, MD5 and MD6 in this catchment show higher channel sediment contributions from T1-T7. These samples corresponded with greater sediment loads suggesting increased inundation of the channel and short term channel bank source availability, for example, stock access degrading channel banks or channel management (during summer months) or increased cumulative flow. The estimated proportion of road contributions was inconsistent at sub-catchment sites but showed a relatively consistent elevation at MD6. Site MD1 reported high road group contributions in T2, T5, T7 and T9, however, the corresponding uncertainty for these predictions were relatively high also.

Despite relatively high spatial resolution source sampling relative to small catchment sizes ($\sim 10 \text{ km}^2$), the variability of tracer values between sources was frequently lower than the variability of single sources. This prevented the discrimination of the intended potential sources ($n=6$) identified at the outset of the investigation (Rowan et al., 2012; Rose et al., 2018). Definition of additional sources (improved dimensionality), such as land use type within each catchment (arable versus grassland), would improve the resolution of source provenance and, in turn, improve the understanding of the interactions between sediment sources and catchment processes, and onward risk assessment and mitigation. Gruszowski et al. (2003) successfully distinguished grassland and arable topsoils using tracers χ_{HF} , χ_{ARM} , IRM_{880} , Fe, Al, Na and Cu. Equivalent tracers measured in this study were, however, not capable of discriminating arable and grassland topsoils and may be attributed to greater arable and grassland crop rotation. Measurement of additional tracers such as soil enzymes and crop-specific compound specific stable isotopes may be useful to provide greater dimensionality (Nosrati et al., 2011; Blake et al., 2012). Despite this, the sediment

fingerprinting approach validated, and are consequently validated by, the catchment characteristics shown by SSC-discharge hysteresis analysis in these study catchments (Sherriff et al., 2016). The confirmation is a significant finding as storm event discharge-suspended sediment hysteresis analysis compared to sediment fingerprinting strategies requires less in terms of analytical sophistication but more in terms of longer-term high resolution river-side monitoring.

3.4 Implications for catchment management

Despite high agricultural intensity (more than 90% utilisation area and high stocking density/crop yields – Sherriff et al., 2015a), source provenance results indicated channel derived sediments were dominant in some settings. These were likely accelerated by field-scale agricultural management such as the presence and configuration of drainage systems which can increase channel flow velocities. These landscape modifications (aimed at reducing excess soil moisture and increasing the utilisable area for agriculture on hillslopes) may occur in combination with bank destabilisation which are also responsible for increased stream energy and consequently bank erosion. Accordingly, mitigation measures designed to reduce flow velocities may dissipate stream power and encourage deposition of entrained sediments. For example, field wetlands positioned at the edge of field or catchment outlets have been shown to retain $0.01 - 6 \text{ t ha}^{-1} \text{ yr}^{-1}$ of sediment and on-farm buffer strips have reported 2 – 50% efficacy (Ockenden et al., 2012; Collins et al., 2018).

Sediment delivery from field topsoils was greater where low ground cover coincided with good hydrological connectivity (consistently in catchments with poor- or moderate soil drainage or sporadically following extreme rainfall events on well-drained soils). Consistently, low risk soils have also found to be a minor contributor to catchment sediments

loads in Brazil (LeGall *et al.*, 2017). Strategies emphasising soil conservation or disconnecting hydrological pathways may contribute to reduced sediment delivery. On-field, prevention of low groundcover periods, for example by retaining crop residues or increasing surface roughness through conservation tillage practices, should be considered to reduce soil erodibility and efficacy of hydrological transport across soil surfaces (Deasy *et al.*, 2010). In grassland fields, prevention of soil structure degradation and over-grazing may reduce soil erosion risk. Measures to reduce hillslope-to-channel sediment connectivity to encourage deposition of entrained sediments, such as abovementioned field wetlands, buffer-strips or unploughed margins are potential sediment mitigation measures (Rickson, 2014),

Management of the public and farm road network to intercept road derived or road transported sediments at road-stream intersections is a source and/or pathway requiring greater study. Although the proportion of road-derived sediments were generally low in the present study, their subsequent elevation with field topsoil contributions has implications for regulations to minimise soiled water transfers from farm roadways to waters by 2021 (European Union, 2017). Similarly to field ditches, roadside ditches can be designed and managed to encourage deposition of associated particulates transported in the flow which consequently reduces sediment delivery to watercourses (Shore *et al.*, 2016). However, installing such features in established agricultural areas may not be practical.

The separation of sediment load into three source groups, defined here in three catchments with contrasting land use and soil drainage combinations, provides a blueprint for sediment transfer vulnerability in agricultural catchments. Expanding the spatial resolution of source provenance shows good consistency of soil erosion and sediment transfer mechanisms within each catchment but also reveals additional processes which are undetectable at the catchment

511 scale. Previous research in the study catchments (Shore et al., 2014; Mellander et al., 2015;
512 2016; Sherriff et al., 2016, Thomas et al., 2016) and elsewhere (Fryirs et al., 2007; Dupas et
513 al., 2015) has confirmed the influence of hydrological connectivity on the delivery of
514 sediment and nutrients at catchment outlets and the importance of their interactions for in-
515 stream ecology (Davis et al., 2018). Identification and mitigation of hillslope sediment losses
516 must target critical source areas to maximise the success and cost-efficiency of mitigation
517 measures (Shore et al., 2013; Thompson et al., 2013; Thomas et al., 2016). Controls on bank
518 erosion of natural and artificial channels are required and have been typically overlooked in
519 agri-environmental policy, at least at a European level (Collins and Anthony, 2008). The
520 potential trade-off between reducing hillslope soil moisture to sustain or increase agricultural
521 production and the initiation and acceleration of erosion at the field edge must be fully
522 considered.

4 Conclusions

The successful multi-proxy partitioning of parent sediment sources (field topsoil, channel and road) indicated contrasting hillslope versus channel influences in the catchment observatories, according to source availability and transport pathways. The main conclusions are:

- The poorly-drained grassland and moderately-drained arable catchments exported overall greater sediment load overall, 828 and 619 tonnes, respectively, than the well-drained arable catchment, 421 tonnes, due to a greater likelihood of sediment connectivity correlated with impeded soil drainage (or lack of infiltration capacity);
- At the catchment outlets, channel, field topsoil and road contributions were 67%, 27% and 4% in the poorly-drained grassland catchment, 53%, 24% and 24% in the well-drained arable catchment and 8%, 82% and 9% in the moderately-drained arable catchment.
- Greater sediment export in the well-drained catchment during three periods (T4, T5 and T12) corresponded with greater contributions from field topsoils, 29% and 24%, respectively, which were attributed to the establishment of surface hydrological connectivity and consequent surface erosion and overland flow transport following extreme rainfall events;
- Spatially, sediment sources were less variable in predominately poorly-drained grassland or moderately-drained arable catchments due to the consistency of flow pathways and respective source availability (poorly-drained grassland catchment – channels, moderately-drained arable catchment – hillslopes);
- Contributions from roads sources were higher (total 24%) where road-stream intersections were more frequent. This effect was seemingly diminished where a

runoff ditch broke this direct connectivity; catchments with ditches at the roadside had lower estimated road sediment contributions (4 and 9%) suggesting that when designed and managed correctly they are a useful management tool to encourage sediment deposition;

In terms of wider application, successful sediment mitigation measures must consider hillslope and riparian areas in intensive agricultural catchments and consider the likely impact of hydrological or land use modifications on downstream sediment loss risk. Emphasis on certain measures according to setting and process is likely to reduce the burden of mitigation required but also likely to target the most vulnerable sediment source areas. Targeted considerations will be necessary to decrease the environmental impacts of intensive agriculture from sediment loss to meet water quality objectives and also offset the likely increases of these impacts as agricultural management is further intensified.

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811 Tables

812 Table 1. Sub-catchment areas and sample collection dates.

Catchment/ Sample location	Area (km ²)	T1	T2	T3	T4	T5	T6	T7	T8	T9	T10	T11	T12	T13	T14
Poorly-drained grass															
PD1	0.75					19-21/02/2013									22/05/2014
PD2	1.76														
PD3	2.64														
PD4	7.76														
PD5	9.81														
PD6	11.88														
Well-drained arable															
WD1	2.07														
WD2	3.30														
WD3	4.10														
WD4	4.23														
WD5	6.36														
WD6	11.16														
Moderately-drained arable															
MD1	1.16														
MD2	2.08														
MD3	3.10														
MD4	4.13														
MD5	6.81														
MD6	9.48														

813 PD: suspended sediment sample locations in the poorly-drained grass catchment, WD: suspended
814 sediment sample locations in the well-drained arable catchment, MD: moderately-drained arable
815 suspended sediment locations in the moderately-drained arable catchment.

816 Table 2. Summary of tracer source data in the poorly-drained grass catchment.

Tracer	Channel banks & ditches		Field topsoils		Road verges & tracks	
	Mean	Std Dev	Mean	Std Dev	Mean	Std Dev
χ_{LF} ($10^{-6}\text{m}^3\text{kg}^{-1}$)	1.46 ^b	0.40	1.92 ^b	2.55	6.43 ^a	3.05
χ_{HF} ($10^{-6}\text{m}^3\text{kg}^{-1}$)	1.43 ^b	0.40	1.78 ^b	2.27	6.24 ^a	2.97
χ_{FD} (%)	2.07 ^b	1.46	4.43 ^a	2.24	5.10 ^{ab}	8.05
χ_{ARM} ($10^{-7}\text{Am}^3\text{kg}^{-1}$)	0.06 ^b	0.03	0.12 ^b	0.18	0.22 ^a	0.12
SIRM ($10^{-5}\text{Am}^3\text{kg}^{-1}$)	248.76 ^b	122.81	168.25 ^c	152.83	1210.55 ^a	717.62
bIRM _{soft} ($10^{-5}\text{Am}^3\text{kg}^{-1}$)	102.34 ^b	46.90	103.83 ^b	116.67	685.86 ^a	373.48
bIRM _{hard} ($10^{-5}\text{Am}^3\text{kg}^{-1}$)	23.38 ^a	6.15	13.95 ^b	4.33	24.54 ^a	13.48
SIRM/ χ_{LF}	139.73 ^b	33.70	80.02 ^c	21.96	182.11 ^a	61.92
SIRM/ χ_{ARM}	3882.79 ^b	2545.56	1577.60 ^c	814.41	5805.23 ^a	3286.54
$\chi_{ARM}/\chi_{LF}^{\dagger}$	0.04	0.01	0.04	0.02	0.03	0.01
H-ratio ($10^{-5}\text{Am}^3\text{kg}^{-1}$)	225.38 ^b	118.11	154.31 ^c	149.03	1186.01 ^a	713.57
Cd* (mg kg^{-1})	0.20	0.11	0.08	0.04	0.18	0.14
Co (mg kg^{-1})	13.45 ^a	2.82	6.17 ^b	1.16	11.85 ^a	3.16
Cr (mg kg^{-1})	23.75 ^a	8.38	17.65 ^b	2.74	22.54 ^a	7.96
Cu (mg kg^{-1})	20.11 ^b	5.96	12.30 ^c	3.89	28.89 ^a	7.36
Mn* (mg kg^{-1})	1233.14	647.86	511.19	211.98	1285.03	464.69
Ni (mg kg^{-1})	24.31 ^a	5.24	12.73 ^b	2.37	22.02 ^a	6.36
Pb (mg kg^{-1})	15.70 ^b	2.91	17.75 ^b	3.74	44.63 ^a	59.61
Zn (mg kg^{-1})	62.57 ^b	12.52	41.35 ^c	6.71	122.07 ^a	41.48
^{234}Th (Bq kg^{-1})	36.94 ^a	14.80	21.65 ^b	5.74	29.75 ^a	10.21
^{235}U (Bq kg^{-1})	47.11 ^a	16.00	29.68 ^b	11.38	50.97 ^a	20.55
^{228}Ac (Bq kg^{-1})	27.22 ^a	7.41	17.02 ^b	4.74	26.55 ^a	8.72
^{137}Cs (Bq kg^{-1})	0.90 ^b	1.60	5.84 ^a	2.45	8.96 ^a	8.05
^{40}K (Bq kg^{-1})	804.67 ^a	13.76	634.55 ^b	89.85	860.75 ^a	140.24
$^{210}\text{Pb}_{\text{unsupp.}}$ (Bq kg^{-1})	10.72 ^b	7.59	10.08 ^b	10.79	60.97 ^a	31.88

817 Tracers removed from analysis: [†]tracer failed Kruskal-Wallis test ($p>0.05$) showing no statistically
818 significant differences in tracer values between source groups; statistically significant differences
819 between source tracer groups (as determined by pairwise Dunn-Bonferroni testing (adjusted $p < 0.05$)
820 are denoted by different letters). * frequent non-conservativeness displayed in river sediment samples.

821 Table 3. Summary of source tracer data in the well-drained arable catchment.

Tracer	Channel banks & ditches		Field topsoils		Road verges & tracks	
	Mean	Std Dev	Mean	Std Dev	Mean	Std Dev
χ_{LF} ($10^{-6}\text{m}^3\text{kg}^{-1}$)	1.81 ^b	1.21	13.33 ^a	8.69	12.51 ^a	2.15
χ_{HF} ($10^{-6}\text{m}^3\text{kg}^{-1}$)	1.73 ^b	1.06	11.87 ^a	7.70	11.70 ^a	2.10
% χ_{FD} (%)	2.28 ^b	1.84	7.31 ^a	0.96	6.39 ^a	1.66
χ_{ARM} ($10^{-7}\text{Am}^3\text{kg}^{-1}$)	0.08 ^b	0.07	0.86 ^a	0.54	0.59 ^a	0.14
SIRM ($10^{-5}\text{Am}^3\text{kg}^{-1}$)	121.15 ^b	57.29	700.69 ^a	484.98	1257.31 ^a	410.67
IRM _{soft} ($10^{-5}\text{Am}^3\text{kg}^{-1}$)	77.53 ^b	45.88	579.05 ^a	420.24	865.06 ^a	242.54
IRM _{hard} ($10^{-5}\text{Am}^3\text{kg}^{-1}$)	13.56 ^b	18.35	12.85 ^{ab}	6.03	17.04 ^a	5.51
SIRM/ χ_{LF}	58.10 ^a	15.51	36.40 ^b	6.94	99.03 ^a	30.20
SIRM/ χ_{ARM}	1532.81 ^a	561.87	545.66 ^{ab}	86.74	2334.86 ^a	1083.26
χ_{ARM}/χ_{LF}	0.03 ^b	0.01	0.05 ^a	0.01	0.05	0.01
H-ratio ($10^{-5}\text{Am}^3\text{kg}^{-1}$)	107.59 ^b	61.23	687.84 ^a	480.56	1240.27 ^a	406.31
Cd (mg kg^{-1})	0.14 ^b	0.08	0.14 ^b	0.07	0.50 ^{ab}	0.43
Co (mg kg^{-1})	16.29 ^a	6.23	9.47 ^b	1.40	12.68 ^a	1.30
Cr (mg kg^{-1})	30.78 ^a	5.34	19.27 ^b	2.88	25.89 ^a	5.69
Cu (mg kg^{-1})	24.36 ^a	7.49	14.75 ^b	2.93	32.13 ^a	14.38
Mn (mg kg^{-1})	969.36 ^b	263.5	1140.24 ^{ab}	247.92	1344.56 ^a	224.16
Ni (mg kg^{-1})	31.72 ^a	6.85	15.55 ^b	2.70	24.20 ^a	3.17
Pb (mg kg^{-1})	20.75 ^b	6.60	17.50 ^b	2.58	32.91 ^a	12.31
Zn (mg kg^{-1})	76.62 ^a	19.79	58.45 ^b	8.66	132.66 ^a	51.62
$^{234}\text{Th}^\dagger$ (Bq kg^{-1})	212.77	60.25	147.07	39.23	173.82	53.14
$^{235}\text{U}^\dagger$ (Bq kg^{-1})	126.63	43.46	102.55	29.92	99.89	47.52
^{228}Ac (Bq kg^{-1})	141.91 ^a	45.13	111.74 ^b	32.02	135.44 ^{ab}	63.69
^{137}Cs (Bq kg^{-1})	19.42 ^b	18.37	92.55 ^a	40.23	126.90 ^a	43.40
^{40}K (Bq kg^{-1})	948.11 ^a	157.55	662.97 ^b	102.46	849.42 ^a	111.10
$^{210}\text{Pb}_{\text{unsupp.}}$ (Bq kg^{-1})	0 ^c	0	11.41 ^b	4.83	32.69 ^a	26.31

822 Tracers removed from analysis: [†]tracer failed Kruskal-Wallis test ($p>0.05$) showing no statistically
823 significant differences in tracer values between source groups; statistically significant differences
824 between source tracer groups (as determined by pairwise Dunn-Bonferroni testing (adjusted $p < 0.05$)
825 are denoted by different letters).

826 Table 4. Summary of source tracer data in the moderately-drained arable catchment.

Tracer (units defined in text)	Channel ditches	banks	&	Field topsoils	Road verges & tracks		
	Mean	Std Dev		Mean	Std Dev	Mean	Std Dev
χ_{LF} ($10^{-6}\text{m}^3\text{kg}^{-1}$)	2.55 ^a	1.62		1.56 ^b	0.67	3.76 ^a	1.49
χ_{HF} ($10^{-6}\text{m}^3\text{kg}^{-1}$)	2.45 ^a	1.46		1.48 ^b	0.61	3.66 ^a	1.48
% χ_{FD} (%)	2.56 ^b	1.66		3.74 ^a	1.44	2.90 ^{ab}	0.95
χ_{ARM}^{\dagger} ($10^{-5}\text{Am}^3\text{kg}^{-1}$)	0.13	0.05		0.13	0.07	0.16	0.04
SIRM ($10^{-5}\text{Am}^3\text{kg}^{-1}$)	339.21 ^a	133.33		177.31 ^b	80.41	510.37 ^a	229.26
IRM _{soft} ($10^{-5}\text{Am}^3\text{kg}^{-1}$)	216.48 ^a	96.64		111.69 ^b	54.78	323.92 ^a	142.46
IRM _{hard} ($10^{-5}\text{Am}^3\text{kg}^{-1}$)	13.38 ^a	4.10		10.70 ^a	2.95	14.21 ^a	5.22
SIRM/ χ_{LF}	144.99 ^a	42.78		92.14 ^b	15.30	123.66 ^a	21.12
SIRM/ χ_{ARM}^*	2569.42	752.73		1100.94	211.93	2880.03	997.97
χ_{ARM}/χ_{LF}	0.06 ^b	0.01		0.07 ^a	0.01	0.04 ^b	0.01
H-ratio ($10^{-5}\text{Am}^3\text{kg}^{-1}$)	325.83 ^a	130.99		166.61 ^b	78.22	496.16 ^a	225.38
Cd* (mg kg ⁻¹)	0.50	0.12		0.40	0.09	0.44	0.08
Co (mg kg ⁻¹)	9.96 ^a	1.21		7.42 ^b	1.11	10.49 ^a	0.77
Cr* (mg kg ⁻¹)	26.88	5.04		22.50	1.90	27.54	3.27
Cu (mg kg ⁻¹)	20.61 ^b	2.98		19.57 ^b	3.60	31.42 ^a	6.66
Mn* (mg kg ⁻¹)	787.22	290.03		486.47	116.21	674.62	87.45
Ni (mg kg ⁻¹)	33.05 ^a	5.60		23.58 ^b	3.15	34.91 ^a	2.55
Pb (mg kg ⁻¹)	35.10 ^{ab}	5.08		30.68 ^b	6.21	37.61 ^a	9.17
Zn (mg kg ⁻¹)	69.60 ^b	12.66		61.40 ^b	10.98	111.54 ^a	29.39
²³⁴ Th [†] (Bq kg ⁻¹)	30.04	9.09		32.67	10.21	27.11	8.00
²³⁵ U [†] (Bq kg ⁻¹)	36.51	6.92		42.81	10.15	43.77	10.78
²²⁸ Ac [†] (Bq kg ⁻¹)	26.46	6.54		23.77	5.01	25.30	4.42
¹³⁷ Cs [†] (Bq kg ⁻¹)	11.54	12.33		8.62	2.67	9.50	4.89
⁴⁰ K (Bq kg ⁻¹)	922.79 ^a	133.77		731.64 ^b	100.58	886.23 ^a	70.27
²¹⁰ Pb _{unSUPP.} (Bq kg ⁻¹)	12.08 ^b	6.30		12.46 ^{ab}	6.32	22.58 ^a	13.78

827 Tracers removed from analysis: [†]tracer failed Kruskal-Wallis test ($p>0.05$) showing no statistically
828 significant differences in tracer values between source groups; statistically significant differences
829 between source tracer groups (as determined by pairwise Dunn-Bonferroni testing (adjusted $p < 0.05$)
830 are denoted by different letters). * frequent non-conservativeness displayed in river sediment samples.

831

832

833 Table 5. Source group discrimination (expressed as percent of original grouped cases correctly
834 classified) of tracer arrays for target suspended sediment samples in the study catchments.

835

Catchment	% of original grouped cases correctly classified	Number of suspended sediment samples
Poorly-drained grassland	89.5	1
	91.9	2
	93.0	2
	94.2	1
	95.3	12
	96.5	6
	97.7	39
Well-drained arable	90.9	4
	93.2	4
	95.5	8
	97.7	46
Moderately-drained grassland	84.2	1
	89.5	2
	91.2	2
	93.0	4
	94.7	9
	96.5	17
	98.2	47

836

837 Figure captions

838 Figure 1. Study catchment locations, outlet coordinates and source and stream sediment sampling
839 locations in the poorly-drained grassland (PD), well-drained arable (WD) and moderately-drained
840 arable catchments (MD).

841

842 Figure 2. Canonical Discrimination Functions (output from Multiple Discriminant Analysis) of the
843 full source sample tracer datasets collected from the, a) poorly-drained grassland, b) well-drained
844 arable, and c) moderately-drained arable catchments categorised by source group.

845

846 Figure 3. Load specific un-mixing of median source predictions of outlet suspended sediment samples
847 in the, a) poorly-drained grass, b) well-drained arable and c) moderately-drained arable catchments
848 over the study period.

849

850 Figure 4. Median source predictions at sub-catchment and outlet locations in the three study
851 catchments. Site names are denoted by catchment abbreviation: PD – poorly-drained grass, WD –
852 well-drained arable and MD – moderately-drained arable with increasing numbers representing sites
853 with increasing sub-catchment areas. Time period, T, represents the period of deployment of time-
854 integrated suspended sediment samplers between May 2012 and May 2014 (see table 1). Grey squares
855 indicate no un-mixed data.

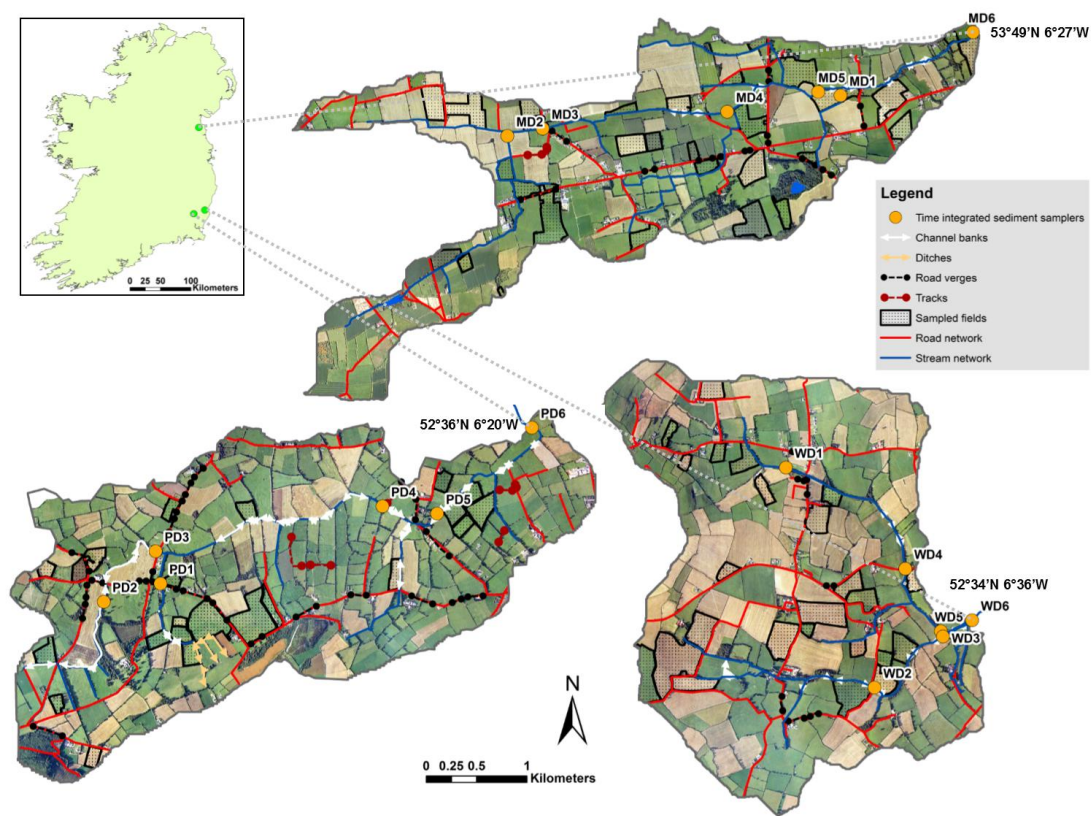
856

857 Figure 5. Combined 95-percentile uncertainty envelopes for sediment source predictions in three
858 study catchments. Site names are denoted by catchment abbreviation: PD – poorly-drained grass, WD
859 – well-drained arable and MD – moderately-drained arable with increasing numbers representing sites
860 with increasing sub-catchment areas. Time period, T, represents the period of deployment of time-

861 integrated suspended sediment samplers between May 2012 and May 2014 (see table 1). Grey squares
862 indicate no un-mixed data.

863 Figures

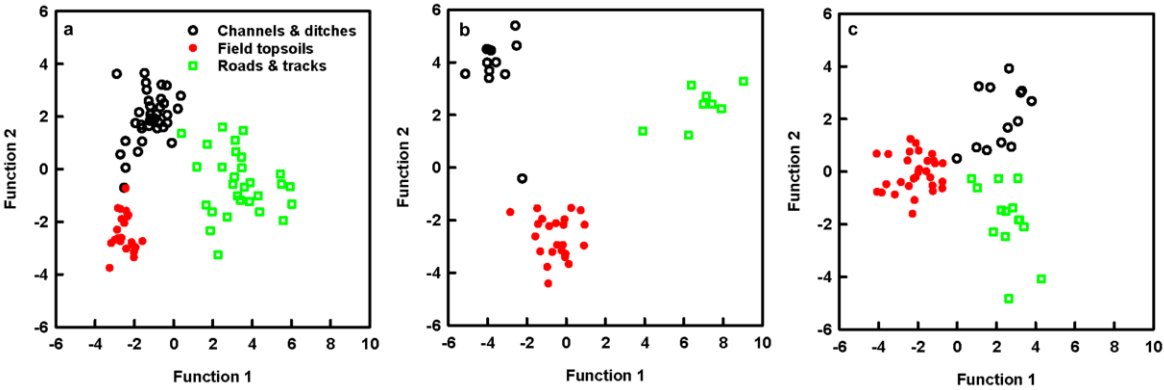
864 Figure 1



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867 Figure 2



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870 Figure 3

